Angular Distributions of Resonant and Non-Resonant Auger Electrons as a Test Case for the Validity of the Spectator Model: The Argon L_2MM Case

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INTRODUCTION

Ever since the first measurements of resonant Auger spectra, following excitation of inner-shell resonances, have been measured they have been compared with normal Auger spectra following above threshold ionisation. A strong similarity was anticipated because the excited electron could be considered as a spectator of the decay process, shielding the core hole and thereby shifting the whole electron spectrum to higher kinetic energies [1]. This simple picture was true for many cases but pronounced deviations from the model were found soon after the study of these processes was tremendously promoted by the use of monochromatic synchrotron radiation.

RESULTS

We have measured the Ar $2p \rightarrow ns,d$ selective excitation with high resolution and a complete analysis can be found in Langer et al. [2]. Here we will only describe the case of the $2p_{1/2} \rightarrow nd$ resonances.

$2p_{1/2} \rightarrow nd$ Resonances.

The complete $2p_{1/2} \rightarrow 3d$ resonant Auger spectrum is shown in Figure 1. Contrary to the $2p \rightarrow 4s$, this resonant transition does not obey the predicted behavior of the spectator model in its most simple form, i.e. the observed spectra are much different from the normal Auger spectra. Instead, the spectra are dominated by shake-up processes. A possible reason for this has been discussed by Aksela and Mursu [3], who have demonstrated the importance of the shake-up process arising from the different degree of collapse of the 3d wave function in the initially excited $2p \rightarrow 3d$ state. This is due to a larger repulsive exchange interaction between the 3p and 3d electrons in the $^1P_1$ state, as compared to the final ionic state. Another explanation has been provided by [4], who has described in some detail the spatial properties of the wave function which leads to a high shake-up probability.

No theoretically predicted β values are available, therefore no comparison is possible at present. However, a comparison with the spectator model can be made if we restrict it to the so-called ”gross spectator model“ (GSM) and to the $2p_{1/2} \rightarrow 3d$ resonance. In the light of the argumentation at the beginning of this paragraph this seems to be unjustified because of the difference in the structures of the resonant and non-resonant spectrum. But, as mentioned earlier, the validity of the spectator model may be discussed in the context of two different situations: the presence of a strong shake-up probability, and the existence of strong configuration interaction and singlet-triplet mixing. It is the second situation that really causes a breakdown of the spectator model [5]. The shake-modified spectator model would predict the same β values for the diagram and shake-up transitions because shake-up probability is basically an overlap factor that does not affect the rest of the Coulomb matrix elements. Thus,

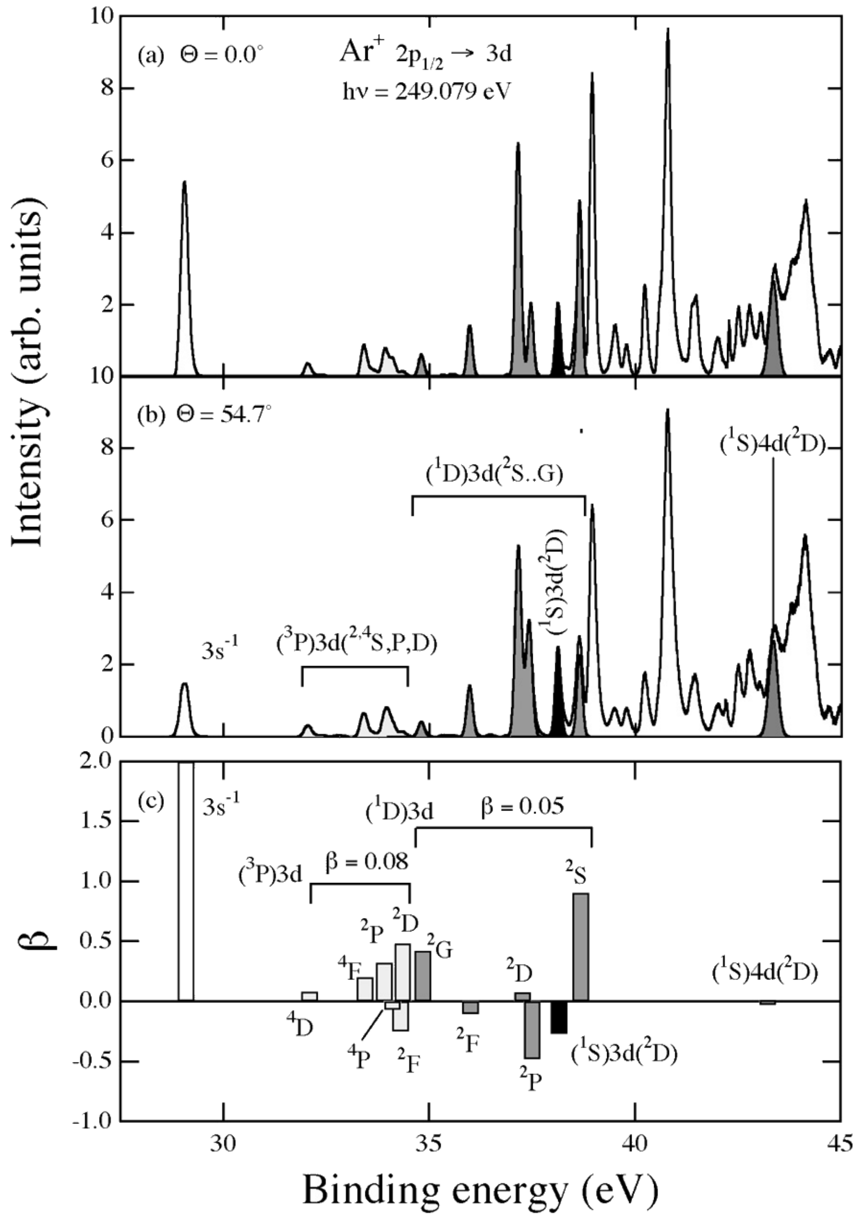

Fig. 1. High resolution resonant Auger spectrum taken on the Ar $2p_{1/2}$ →3d resonance. The light-grey shaded lines correspond to the ^{3}P core, the dark-grey shaded lines to the ^{1}D core, and the black lines to the ^{1}S core. The β values in part (c) are shown by bars shaded correspondingly. The average β of all lines with the same core parent term are shown above or underneath by their numbers

the spectrum may look quite different but the angular distribution is similar. How could this be proved without numerical calculations? The test case are the $2p_{1/2}$→nd excitations because they converge to the same ionisation limit as the ns excitations with a spherical $2p_{1/2}$ final ionic state. Therefore, all the arguments for the ns excitations apply also to the nd excitations. Since we have no β values for the different multiplets we can compare our data only with the sum of the doublet and quartet states. This sum of β has to be zero according to the spectator model which in this particular restricted version is known as gross spectator model. A comparison with the ^{1}S, ^{1}D and ^{3}P states shows β values of -0.27, 0.05, and 0.08, respectively, corresponding quite well - except for the ^{1}S state - with the prediction of the spectator model.

The deviation of the ^1S core state is most pronounced for the 3d ^2D state with its β = -0.27. The even stronger shake-up state 4d ^2D, however, shows already a β value close to zero in accordance with the gross spectator model. The collapsed 3d wavefunction may cause mixing between different parent states but also between the $2p_{1/2}$ and $2p_{3/2}$ core hole states. Both interactions would cause a measurable deviation from the GSM predictions. These deviations, however, seem to disappear already for n=4 suggesting to consider the 3d behavior as a collapse induced anomaly rather than a common deviation in resonant Auger decay. In order to prove if this statement holds also for higher n, the spectrum of the $2p \rightarrow 4d$ resonance was examined. Concentrating on the ^1S, ^1D and ^3P parent states with 4d spectator electron the sum of the β parameters are the following: 0.06, 0.11, and -0.04. Obviously, the correspondence with the gross spectator model prediction is much more favorable than in the case of the $2p_{1/2} \rightarrow 3d$ excitation. All three parent states of the 4d spectator electron still agree quite well. In contrast, the 3d shake-down state deviates significantly from the predicted vanishing Auger anisotropy. The observed β value is somewhat more negative than the value observed for the 3d excitation, but still relative similar.

In summary, we have reported [2] on new measurements of the resonant Auger spectra following selective excitation of the Ar $2p \rightarrow ns,d$ resonances by angle-resolved electron spectroscopy. The results are interpreted in terms of testing the validity of the spectator model in its different forms such as gross and strict spectator model, in particular in the case of the $2p_{1/2}$ excitations which are specifically suited for testing the spectator model due to alignment considerations. The comparison proves the validity of both spectator models for the Ar $2p \rightarrow 4s$ but shows the breakdown of the strict but also a partial breakdown of the gross spectator model for the Ar $2p \rightarrow nd$ excitations for n=3. Mixing of the core and maybe hole states due to the 3d wave function collapse is considered the most probable reason for this breakdown. The recovery of the gross spectator model, however, starts unexpectedly early with n=4, a trend being experimentally confirmed via shake-up states up to n=6. Specific measurements are suggested to prove this statement for even higher n up to threshold.

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REFERENCES

1. M. O. Krause, T. A. Carlson, and W. E. Moddemann , J. Phys. (Paris) **32** C4-139 (1971).
2. B. Langer, N. Berrah, A. Farhat, M. Humphrey, D. Cubaynes, A. Menzel and U. Becker, J. Phys. B **30**, 4255 (1997).
3. H. Aksela and J. Mursu, Phys. Rev. A **54** 2882 (1996).
4. M. Meyer, E. v. Raven, B. Sonntag, and J.E. Hansen, Phys. Rev. A **43** 177-188 (1991).
5. U. Hergenhahn, S.B. Whitfield, J. Tulkki, F. Heiser, N.M. Kabachnik, B. Langer, and U. Becker, Phys. Rev. A **55** 2050-2066 (1996).

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